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ULTRAVIOLET RAY INTERCEPTING AGENT  
(Shigai sen shadan zai)

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## Specification

### 1. Title of Invention

Ultraviolet ray intercepting agent

### 2. Scope of Patent Claims

(1) An ultraviolet ray intercepting agent is made from a complex oxide of zinc and more than 1 type of the following metals selected from the group such as aluminum, iron, chromium, cerium, zirconium and titanium. The average particle size is 0.001 micrometer - 0.5 micrometer.

### 3. Detail explanation of the invention

(Industrial field of use)

The invention pertains to an ultraviolet ray intercepting agent consisting of a complex oxide powder of a specific metal having infrared ray and visible rays intercepting function. The ultraviolet ray that can be interrupted is in the wavelength of 260 nm - 400 nm.

(Prior Art)

Since the ultraviolet intercepting agent can interrupt the ultraviolet ray, absorbs or dispersed the ultraviolet rays, it is known that the intercepting function due to the metal oxide powder has this ratio of particle of

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<sup>1</sup> the numbers in the margin indicate pagination in foreign text

powder/radiation wavelength equal to  $\frac{1}{2}$ . The examples of the metal oxide powder used are such as titanium oxide, zinc oxide and iron oxide and the organic acids used are such as salicylic acid, para alumino benzoic acid, silicic acid and the ester group or the benzophenone group of these.

The metal oxide powder is used as the catalyst for the oxidation reaction. An example of that production method is the addition of ammonia water or urea to the chloride and sulfate of the metal and hydrolyzed. The baking method for the hydroxide substance that is obtained is that the isopropoxide of the titanium and the zinc sulfate are hydrolyzed respectively, the  $\text{TiO}_2 \cdot n\text{H}_2\text{O}$  and the  $\text{Zn}(\text{OH})_2$  are mixed and baked, this method was disclosed in ("Catalyst" Vol. 19, No. 5, 1977, page 350 - 352, Catalyst Society Journal).

Also, the mixture of alkoxide of the silicon element and titanium are hydrolyzed. That hydrolyzed product is baked and it is known that this product is used as the additive for cosmetics having the ultraviolet ray intercepting function (Patent Publication No. 59 - 227813).

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In addition, a method for obtaining the fine particles is the method for performing the oxidation decomposition, hydrolyzing with steam or oxygen, the titanium chloride

particles obtained have particle size in this range, 0.002 micrometer - 0.05 micrometer.

In the zinc oxide, the method used is the vapor phase oxidation for vaporizing the zinc metal. Thus, the particle size obtained is in the range of 0.5 micrometer - 1 micrometer.

(The problems resolved by the invention)

The zinc oxide powder, titanium oxide powder and iron oxide powder obtained in such a method displayed good ultraviolet intercepting properties when selected with the above particle size but it is lacking in intercepting the visible rays and the infrared rays. In addition, the ultraviolet rays interception function is insufficient with large particle size with the lacking in intercepting the visible rays and the infrared rays.

Also, there is the problem with these metal oxide powder, it has oxidation catalytic function, the coexisting organic substance is modified.

Several metal salts are hydrolyzed with the ammonia water in aqueous solution. In the method for obtaining the complex oxide, the particle size of the complex oxide is not uniformed. The ultraviolet rays interception function is sufficient but the good permeation of the visible rays and the infrared rays cannot be obtained.

(The means for resolving the problems)

By focusing on the above problems, the inventors found an ultraviolet ray intercepting agent that has good ultraviolet ray interception function, the permeation of the visible ray and infrared rays is large. Also, there is no problem with the modification of the coexisting organic substance. As a result, the invention attained success.

That is, the invention offers an ultraviolet ray intercepting agent that is made from a complex oxide of zinc and more than 1 type of the following metals selected from the group such as aluminum, iron, chromium, cerium, zirconium and titanium. The average particle size is 0.001 micrometer - 0.5 micrometer.

The invention is explained in further detail below.

The examples of the ultraviolet ray intercepting agent in the invention are such as the complex oxide of the zinc (it is referred to as Zn below) and the metal of more than 1 type of metal selected from these groups, aluminum, iron, chromium, cerium, zirconium and titanium (refer to below as Al, Fe, Cr, Ce, Zr, Ti). The specific examples are the two components complex oxides such as Zn - Al, Zn - Fe, Zn - Cr, Zn - Ce, Zn - Zr, Zn - Ti; the three components complex oxides are such as Zn - Al - Fe,.....Zn - Zr - Ti. The four components complex oxides are such as Zn - Al - Fe

- Cr,.....Zn - Cr - Ce - Ti; the five components complex oxides are such as Zn - Al - Fe - Cr - Ce,.....Zn - Fe - Cr - Zr - Ti. The six components complex oxides are such as Zn - Al - Fe - Cr - Ce - Zr,.....Zn - Fe - Cr - Ce - Zr - Ti and the 7 components complex oxides is Zn - Al - Fe - Cr - Ce - Zr - Ti. /3

It is preferred that these complex oxides are used alone but of course it is most preferred that 2 or more of these types are mixed together.

In this implementation of the invention, the constitution combination of the other metals selected from this group - Ti, Al, Fe, Cr, Ce, Zr and zinc constituting the complex oxides is different depending on the metal. For example, the atomic ratio of the combination of zinc and titanium, that is, the titanium to Zinc 1 is 0.01 - 0.25 but preferably 0.05 - 0.1. The aluminum to zinc 1 for the zinc and aluminum is 0.001 - 0.5, preferably 0.01 - 0.4. For the zinc and iron, the iron to zinc 1 is 0.0001 - 0.5, preferably 0.0001 - 0.2. For zinc and chromium, the chromium to zinc 1 is 0.0005 - 0.5, preferably 0.0005 - 0.3. For the zinc and cerium, the cerium to zinc 1 is 0.00005 - 2 but preferably 0.00005 - 0.5. For zinc and zirconium, zirconium to zinc 1 is 0.0005 - 0.5 but preferably 0.0005 - 0.3.

When the constitutional combination of the metal to the zinc 1 atomic ratio is outside the above range, the ultraviolet ray intercepting function deteriorates. If the atomic ratio of zinc 1 to iron exceeds 0.5, since the permeation of the visible ray and infrared ray is reduced, this is not desirable.

Also, the addition amount of the metal in the complex oxide made from zinc and several metals is the various addition amount ranges for the zinc and other metals in two component system. A preferred range of various metals are used in the two component system of zinc and other metals.

For the complex oxide in the invention, the metals used for the reaction of more than 1 types selected from Al, Fe, Cr, Ce, Zr, Ti and zinc used as the original raw materials. These are essential to form the complex oxides. The complex oxide is formed with little by products. That product has ultraviolet intercepting function of above 90% and visible ray and infrared ray intercepting function of above 60%. It is preferred this complex oxide consists one part of metal oxide.

The complex oxide for the ultraviolet intercepting agent of the invention has the constitution of bonding to the metal via the oxygen atom.



For example, since the zinc atom consists of a bond to other metals via the oxygen atoms, for example, - Zn - O - Ti <, Zn - .....>Cr - O- Zn - O - Zr, it is preferred that all of the Zn atoms bond with the other metals via the above described oxygen. This structure can be recognized by the infrared spectral according to JISK0117.

The Zn atom consists of these bonds R - O - Zn - O - RA, - Zn - O - RA (R is the metal atoms selected from these groups Al, Fe, Zr, Ti, Ce, Cr, A is the bonding means, R is 1 valent). For the purpose of the invention, it has excellent ultraviolet ray intercepting function. The powder that is good in the visible ray and infrared ray intercepting cannot be obtained.

The size of the ultraviolet ray of the invention has an average size of below 0.5 micrometer but preferably below 0.2 micrometer.

As the particle size is large, the ultraviolet ray intercepting capability is low. In addition, when used in cosmetics, it crumbles easily or the dispersion is poor when used in the film for agriculture application or a film for food packaging.

On the other hand, when the particle size is too small, the ultraviolet ray intercepting function is sufficient but it is difficult to handle and the oxidation

catalytic function cannot be realized. The particle size is in the range of above 0.001 micrometer but preferably in the range of above 0.005 micrometer.

An example of this production method of this type of complex oxide is that the zinc salt and 1 type or 2 types of metal salt selected from these, zirconium salt, iron salt, chromium salt, cerium salt, aluminum salt and titanium salt are selected from urea and hexamethylene tetramine in the alcohol aqueous solution. The hydrolysis is performed under the presence of at least 1 type of hydrolysis sedimentation agent. Next, a baking method is used for baking the hydrolyzed product. /4

Here, the raw material for the zinc salt, the aluminum salt, the iron salt, the chromium salt, the cerium salt, the zirconium salt and the titanium salt are the inorganic or organic salt that are soluble in water. For example, chloride, sulfate, nitrate and acetate.

The example of the aluminum salt is the polyvinyl chloride aluminum can be used.

Also, titanium alkoxide can be used instead of titanium salt.

Next, the alcohol aqueous solution is above 5 % content at the alcohol concentration but preferably above 10 % content.

If the alcohol concentration is below 5 % content, the average particle size of the complex oxide produced is below 0.5 micrometer. The particle distribution is wide, a large particle size is mixed in which is undesirable.

In addition, when the alcohol is in excess, the effect desired cannot be obtained.

An example of the type of alcohol is the solubility in water which is above 1% content. For example, the monovalent alcohol such as methanol, ethanol, isopropanol, normal propanol and the multivalent alcohol group such as ethylene glycol, diethylene glycol, propylene glycol and glycerin.

The hydrolysis sedimentation agent is urea and hexamethylene tetramine or a mixture of both of these is used as the aqueous solution.

The amount of the hydrolysis sedimentation agent is more than 2 times the theoretical amount required to neutralize the acid produced from the hydrolyzing the metal salt, the preferred amount is 3 - 5 times the amount.

The hydrolysis reaction is performed at a normal temperature to 100 deg C under agitation, it is preferred that this is performed above 70 deg C for 3 - 6 hours. After the reaction has ended, the pH of the reaction solution rises, it is recognized to be close to 7.

The hydrolyzed product is filtered and removed with a solid - liquid separation operation using a decantation method. It is washed then dried sufficiently from room temperature to 200 degree C. It is baked for 1 - 3 hours at the temperature range of 350 deg C - 1100 deg C.

After baking, the particles precipitated twice are pulverized (broken). The pulverizing is performed using hammer mill, pore mill, an agitator, a vibrating mill, etc.

A specific application of the ultraviolet ray intercepting agent of the invention is the additive used in cosmetics, the film used in food packaging, agriculture, paint, and food containers. The ultraviolet ray intercepting agent used in the invention can be combined with a commonly known ultraviolet ray intercepting agent such as titanium oxide, zinc oxide and iron oxide.

(Effect of Invention)

The ultraviolet ray intercepting agent made from a complex oxide of particle size 0.001 micrometer - 0.5 micrometer of the invention has an ultraviolet ray intercepting capability of above 90 % for rays having wavelengths of below 400 nm. The permeability of the visible ray and infrared ray is above 60 %. (wavelength of 400 nm - 3000 nm). The catalyst activity is almost non existence. Since the coexisting organic substance is not

modified, the ultraviolet ray intercepting agent can be used in various fields, it is a valuable product in the industry.

(Implementation example)

The ultraviolet ray intercepting agent of the invention is explained based on the implementation examples and comparison examples. The implementation example show one implementation state of the invention. The range of the invention is not particularly restricted.

Furthermore, each measurement methods used in the implementation example and comparison example of the invention are given below.

Metal atom ratio: atomic ray absorption analysis method

Particle size of oxide: scanning electronic microscope photograph method

Atom bonding state in the complex oxide: Complex oxide powder of 0.03 g is mixed with low density polyethylene powder of 0.47 g, a film of thickness of 50 micrometers is made, it was mixed and fused with 2 rollers (temperature of 100 deg. C - 160 deg C) /5

A low density polyethylene film of thickness of about 50 micrometers containing no complex oxide is used as the comparison film.

The infrared spectral is measured according to JISK0117 for both films, it is analyzed from the specific absorption.

Spectral transmissivity:

The bonding state of the above complex oxide is checked. It is measured according JISK0115.

Implementation example 1

0.45 g of 97% of aluminum nitrate used in the industry and 36.2 g of 95 % of zinc nitrate used in industry are introduced into a 1200 ml of ethanol. This is agitated, mixed and dissolved in 30 minutes.

Next, 283 g of 99% of hexamethylene tetramine is added to the aqueous solution dissolved in 1600 ml of water. In addition, it is agitated.

This is heated, it is heated to a temperature of 80 deg C and agitated for 5 hours and heated to 100 deg C for 1 hour, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried to 150 deg C and pulverized with a vibration mill.

Next, it is baked for 1 hour at a temperature of about 700 deg C, 91.8 g of the fine powder of average particle size of the powder is 0.01 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Al of 100/0.1.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 2

44.7 g of 97% of aluminum nitrate used in the industry and 362 g of 95 % of zinc nitrate 6 used in industry are introduced into a 1200 ml of ethanol. This is agitated, mixed and dissolved in 30 minutes.

Next, 283 g of 99% of hexamethylene tetramine is added to the aqueous solution dissolved in 1600 ml of water. In addition, it is agitated.

This is heated, it is heated to a temperature of 80 deg C and agitated for 5 hours and heated to 100 deg C for 1 hour, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried to 150 deg C and pulverized with a vibration mill.

Next, it is baked for 1 hour at a temperature of about 700 deg C, 97.1 g of the fine powder of average particle size of the powder is 0.01 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Al of 100/0.1.

The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Al - < bond}$  (special absorption of  $650 \text{ cm}^{-1}$ ) is recognized.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 3

193 g of 97% of aluminum nitrate used in the industry and 222 g of 99 % of zinc 2 nitrate aqueous solution used in industry are introduced into a 1200 ml of ethanol. This is agitated, mixed and dissolved in 30 minutes. Next, 367 g of 99% of hexamethylene tetramine is added to the aqueous solution dissolved in 1600 ml of water. In addition, it is agitated. This is heated, it is heated to a temperature of  $80 \text{ deg C}$  and agitated for 5 hours and heated to  $100 \text{ deg C}$  for 1 hour, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried to  $150 \text{ deg C}$  and pulverized with a vibration mill.

Next, it is baked for 1 hour at a temperature of about  $700 \text{ deg C}$ , all the particle size is not larger than 1 micrometer, 103 g of the fine powder of average particle size of the powder is 0.05 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of  $\text{Zn/Al}$  of 100/50.



The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Al - < bond}$  (special absorption of  $650 \text{ cm}^{-1}$ ) is recognized.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 4

2.19 g of titanium tetrachloride and 362 g of 95 % of zinc 6 nitrate aqueous solution are introduced into a 1200 mg of n-propanol. This is agitated, mixed and dissolved in 30 minutes.

Next, 250 g of urea used in industry and 1600 ml of water are added and mixed.

This is heated to a temperature of  $90 \text{ deg C}$  for 4 hours and heated to  $100 \text{ deg C}$  for 1 hour, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained. /6

This is filtered and washed with 1000 ml of water. It is dried to  $150 \text{ deg C}$  and pulverized with a vibration mill.

Next, it is baked for 1 hour at a temperature of about  $850 \text{ deg C}$ , all the particle size is not larger than 1 micrometer, 92.7 g of the fine powder of average particle size of the powder of 0.08 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of  $\text{zn/Ti}$  of 100/1.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 5

21.9 g of titanium tetrachloride and 362 g of 95 % of zinc nitrate aqueous solution are introduced into a 1200 ml of n-propanol. This is agitated, mixed and dissolved in 30 minutes.

Next, 250 g of urea used in industry and 1600 ml of water are added and mixed.

This is heated to a temperature of 90 deg C for 4 hours and heated to 100 deg C for 1 hour, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried to 150 deg C and pulverized with a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 1 micrometer, 101 g of the fine powder of average particle size of the powder of 0.2 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Ti of 100/10.

The atom bonding state of the powder that is obtained is measured, -Zn - O - Ti - < bond (special absorption of 7400 cm-1) is recognized.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 6

32.9 g of titanium isopropoxide made by Nihon Co. and 362 g of 95 % of zinc nitrate aqueous solution are dissolved into a 1200 ml of n-propanol. This is agitated, mixed and dissolved in 30 minutes.

Next, 278 g of urea is dissolved in 1600 ml of water and mixed.

This is heated to a temperature of 90 deg C and agitated for 5 hours, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried to 150 deg C and pulverized with a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 0.5 micrometer, 101 g of the fine powder of average particle size of the powder of 0.1 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Ti of 100/10. The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Ti -}$  bond is recognized.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 7

65.8 g of titanium isopropoxide made by Nihon Co. and 362 g of 95 % of zinc 6 nitrate aqueous solution are dissolved into a 1200 ml of iso-propanol. This is agitated, mixed and dissolved in 30 minutes.

Next, 278 g of urea is dissolved in 1600 ml of water and mixed.

This is heated to a temperature of 80 deg C and agitated for 5 hours, then, it is heated for 1 hour at 100 deg C, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried to 150 deg C and pulverized with a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 0.5 micrometer, 110 g of the fine powder of average particle size of the powder of 0.1 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Ti of 100/20.

The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Ti - < bond}$  (special absorption of 740  $\text{cm}^{-1}$ ) is recognized.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 8

0.052 g of Iron 2 nitrate of 99% industrial strength and 392 g of 95 % industrial strength of zinc 6 nitrate aqueous solution are introduced into a 1200 ml of normal propanol. This is agitated, mixed and dissolved in 30 minutes. Next, 262 g of urea is dissolved in 1600 ml of water and mixed. This is heated to a temperature of 90 deg C and agitated for 4 hours, then, it is heated for 1 hour at 100 deg C, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill. /7

Next, it is baked for 1 hour at a temperature of about 600 deg C, all the particle size is not larger than 1 micrometer, 97.4 g of the fine powder of average particle size of the powder of 0.06 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Fe of 100/0.01.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 9

Similar to Implementation example 8, the raw material iron chloride of 51 g and 302 g of urea are used, the rest

of the example uses the same process. All the particle size is not larger than 1 micrometer, 110 g of the fine powder of average particle size of the powder of 0.08 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Fe of 100/0.01.

The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Fe - < bond}$  (special absorption of  $530 \text{ cm}^{-1}$ ) is recognized.

The spectral transmissivity measurement result is shown in figure 2.

#### Implementation example 10

392 g of Zinc 6 nitrate of 95% industrial strength and 0.027 g of cerium 6 nitrate 1 aqueous solution are introduced into a 1200 ml of iso-propanol. This is agitated, mixed and dissolved in 30 minutes. Next, 262 g of urea of industrial strength is dissolved in 1600 ml of water and mixed. This is heated to a temperature of  $90 \text{ deg C}$  and agitated for 4 hours, then, it is heated for 1 hour at  $100 \text{ deg C}$ , the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at  $150 \text{ deg C}$  and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 600 deg C, all the particle size is not larger than 1 micrometer, 97.5 g of the fine powder of average particle size of the powder of 0.01 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Ce of 100/0.005.

The spectral transmissivity measurement result is shown in figure 2.

#### Implementation example 11

392 g of Zinc 6 nitrate of 95% industrial strength and 0.055 g of cerium 6 nitrate 1 aqueous solution are introduced into a 1200 ml of iso-propanol. This is agitated, mixed and dissolved in 30 minutes. Next, 262 g of urea of industrial strength is dissolved in 1600 ml of water and mixed. This is heated to a temperature of 90 deg C and agitated for 4 hours, then, it is heated for 1 hour at 100 deg C, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 600 deg C, all the particle size is not larger than 1

micrometer, 97.5 g of the fine powder of average particle size of the powder of 0.01 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Ce of 100/0.01.

The spectral transmissivity measurement result is shown in figure 2.

#### Implementation example 12

Similar to Implementation example 11, the raw material zinc chloride of 72.1 g and 200 g of cerium chloride are used, the rest of the example uses the same process. All the particle size is not larger than 1 micrometer, 97.1 g of the fine powder of average particle size of the powder of 0.03 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Ce of 100/200.

The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Ce -}$  bond (a special absorption of  $410 \text{ cm}^{-1}$ ) is recognized.

The spectral transmissivity measurement result is shown in figure 2.

#### Implementation example 13

392 g of Zinc 6 nitrate of 95% industrial strength and 0.264 g of chromium 9 nitrate 2 aqueous solution are introduced into a 1200 ml of normal propanol. This is



agitated, mixed and dissolved in 30 minutes. Next, 266 g of hexamethylene tetramine of industrial strength is dissolved in 1600 ml of water and mixed. This is heated to a temperature of 90 deg C and agitated for 5 hours, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 600 deg C, all the particle size is not larger than 0.05 micrometer, 97.6 g of the fine powder of average particle size of the powder of 0.01 micrometers are obtained. /8

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Cr of 100/0.05.

The spectral transmissivity measurement result is shown in figure 2.

#### Implementation example 14

392 g of Zinc 6 nitrate of 95% industrial strength and 0.527 g of chromium 9 nitrate 2 of 95% industrial strength aqueous solution are introduced into a 1200 ml of normal propanol. This is agitated, mixed and dissolved in 30 minutes. Next, 266 g of hexamethylene teramine of 99% industrial strength is dissolved in 1600 ml of water and

mixed. This is heated to a temperature of 90 deg C and agitated for 5 hours, the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 600 deg C, all the particle size is not larger than 0.05 micrometer, 97.6 g of the fine powder of average particle size of the powder of 0.01 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Cr of 100/0.1.

The spectral transmissivity measurement result is shown in figure 2.

#### Implementation example 15

Similar to Implementation example 14, the raw material zinc chloride of 313 g and 211 g of chromium chloride, 372 g of hexamethylene tetramine are used, the rest of the example uses the same process. All the particle size is not larger than 1 micrometer, 126 g of the fine powder of average particle size of the powder of 0.04 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Cr of 100/50.

The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Cr - < bond}$  (a special absorption of  $610 \text{ cm}^{-1}$ ) is recognized.

The spectral transmissivity measurement result is shown in figure 2.

#### Implementation example 16

392 g of Zinc 6 nitrate of 95% industrial strength and 0.201 g of Zirconium 8 oxychloride aqueous solution are introduced into a 500 ml of normal propanol. This is agitated, mixed and dissolved in 30 minutes. Next, 225 g of urea of industrial strength is dissolved in 2300 ml of water and mixed. This is heated to a temperature of  $80 \text{ deg C}$  and agitated for 5 hours, then, it is heated for 1 hour at  $100 \text{ deg C}$ , the hydrolysis reaction is performed, a hydrolyzed reaction product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at  $150 \text{ deg C}$  and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about  $650 \text{ deg C}$ , all the particle size is not larger than 0.5 micrometer, 98.1 g of the fine powder of average particle size of the powder of 0.01 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of  $\text{Zn/Zr}$  of 100/0.05.

The spectral transmissivity measurement result is shown in figure 3.

#### Implementation example 17

Similar to Implementation example 16, the raw material zirconium chloride of 4.03 g is used, urea is used as the hydrolysis precipitation agent and 310 g of hexamethylene tetramine are used, the rest of the example uses the same process. All the particle size is not larger than 0.5 micrometer, 101 g of the fine powder of average particle size of the powder of 0.01 micrometers are obtained. The composition of this powder product is analyzed and the result is an atom ratio of Zn/Zr of 100/1.

The spectral transmissivity measurement result is shown in the figure.

#### Implementation example 18

Similar to Implementation example 16, the raw material zinc chloride of 313 g is used, 270 g of urea and 129 g of zirconium chloride are used, the rest of the example uses the same process. All the particle size is not larger than 0.5 micrometer, 127 g of the fine powder of average particle size of the powder of 0.02 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Zr of 100/40.

The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Zr - < bond}$  (a special absorption of  $640 \text{ cm}^{-1}$ ) is recognized.

The spectral transmissivity measurement result is shown in the figure 3.

Implementation example 19

18.7 g of titanium tetrachloride is dissolved in 152 g of an aqueous solution of aluminum 9 nitrate of 97% industrial strength and 308 g of zinc 6 nitrate of 95% is dissolved in 200 ml of ethylene glycol. /9

An aqueous solution containing 319 g of Urea that is dissolved in 2600 ml of water is added to this, these are mixed together.

This is heated to  $100 \text{ deg C}$  and the hydrolysis reaction is carried out for 3 hours under agitation, a hydrolyzed product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at  $150 \text{ deg C}$  and pulverized with a vibration mill.

Next, it is baked for 1 hour at a temperature of about  $850 \text{ deg C}$ , all the particle size is not larger than 1 micrometer, 108 g of the fine powder of average particle size of the powder of 0.2 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of  $\text{Zn/Al/Ti}$  of 100/40/10.

The atom bonding state of the powder that is obtained is measured, -Ti - O - Zn - O - Al< bond (a special absorption of 650 cm<sup>-1</sup>, 740cm<sup>-1</sup>) is recognized.

The spectral transmissivity measurement result is shown in figure 3.

Implementation example 20

21.9 g of titanium tetrachloride of industrial strength, 2.06 g of cerium 8 sulfate 1 and 362 g of zinc 6 nitrate of 95% industrial strength are dissolved in 1200 ml of diethylene glycol. This is agitated for 30 minutes and mixed well.

Next, it is mixed with an aqueous solution containing 1600 ml of water and 252 g of industrial strength urea, these are mixed together.

This is heated to 100 deg C and the hydrolysis reaction is carried out for 3 hours under agitation, a hydrolyzed product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 0.5 micrometer, 104 g of the fine powder of average particle size of the powder of 0.02 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Ce/Ti of 100/0.5/10.

The atom bonding state of the powder that is obtained is measured, -Zn - O - Ti< bond (a special absorption of 600 cm-1, 740 cm-1) is recognized.

The spectral transmissivity measurement result is shown in figure 1.

#### Implementation example 21

44.7 g of an aqueous solution of aluminum nitrate of 97% industrial strength, 4.72 g of iron 9 nitrate 2 and 362 g of zinc 6 nitrate of 95% industrial strength are dissolved in 1200 ml of ethanol. This is agitated for 30 minutes and mixed well. Next, it is mixed with an aqueous solution containing 1600 ml of water and 286 g of 99% industrial strength hexamethylene tetramine, these are mixed together and agitated further.

This is heated under agitation, the heating is carried out for 5 hours at 80 deg C. Next, it is heated to 100 deg C for 1 hour, a hydrolyzed product is obtained. This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 700 deg C, all the particle size is not larger than 1

micrometer, 98.5 g of the fine powder of average particle size of the powder of 0.01 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Al/Fe of 100/10/1. The atom bonding state of the powder that is obtained is measured, -Zn - O - Al< bond (a special absorption of 650. cm-1) is recognized.

The spectral transmissivity measurement result is shown in figure 3.

#### Implementation example 22

44.7 g of an aqueous solution of aluminum nitrate of 97% industrial strength, 5.02 g of cerium 6 nitrate 1 and 362 g of zinc 6 nitrate of 95% industrial strength are dissolved in 1200 ml of ethylene glycol. This is agitated for 30 minutes and mixed well.

Next, it is mixed with an aqueous solution containing 1600 ml of water and 286 g of 99% industrial strength hexamethylene tetramine, these are mixed together and agitated further.

This is heated under agitation, the heating is carried out for 3 hours at 100 deg C. A hydrolyzed product is obtained. This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.



Next, it is baked for 1 hour at a temperature of about 700 deg C, all the particle size is not larger than 0.5 micrometer, 98.7 g of the fine powder of average particle size of the powder of 0.02 micrometers are obtained.

The atom bonding state of the powder that is obtained is measured,  $\text{-Zn - O - Al-}$  bond (a special absorption of 650  $\text{cm}^{-1}$ ) is recognized. /10

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Al/Ce of 100/10/1.

The spectral transmissivity measurement result is shown in figure 3.

#### Implementation example 23

21.9 g of titanium tetrachloride of industrial strength, 4.72 g of iron 9 nitrate 2 of 99% industrial strength and 362 g of zinc 6 nitrate of 95% industrial strength are dissolved in 1200 ml of normal propanol. This is agitated for 30 minutes and mixed well.

Next, it is mixed with an aqueous solution containing 1600 ml of water and 253 g of industrial strength urea, these are mixed together and agitated further.

This is heated under agitation, the heating is carried out for 5 hours at 90 deg C. A hydrolyzed product is obtained. This is filtered and washed with 1000 ml of

water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 0.5 micrometer, 104 g of the fine powder of average particle size of the powder of 0.03 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Fe/Ti of 100/1/10.

The atom bonding state of the powder that is obtained is measured, -Zn - O - Ti< bond (a special absorption of 600 cm<sup>-1</sup>, 740 cm<sup>-1</sup>) is recognized.

The spectral transmissivity measurement result is shown in figure 4.

#### Implementation example 24

48.3 g of aluminum nitrate of 97% industrial strength, 5.1 g of iron 9 nitrate 2 of 99% industrial strength, 277 g of zinc 2 acetate of 99% industrial strength and 2.24 g of cerium 8 sulfate 1 are dissolved in 300 ml of ethanol. This is agitated for 30 minutes and mixed well.

Next, it is mixed with an aqueous solution containing 2500 ml of water and 264 g of industrial strength urea, these are mixed together and agitated further.

This is heated under agitation, the heating is carried out for 5 hours at 80 deg C. Then, it is heated to 100 deg C for 1 hour. A hydrolyzed product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 750 deg C, all the particle size is not larger than 0.5 micrometer, 110 g of the fine powder of average particle size of the powder of 0.01 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Al/Fe/Ce of 100/10/1/0.5.

The atom bonding state of the powder that is obtained is measured, -Zn - O - Ti< bond (a special absorption of 650 cm<sup>-1</sup>) is recognized.

The spectral transmissivity measurement result is shown in figure 4.

#### Implementation example 25

152 g of aluminum 9 nitrate of 97% industrial strength, 2.01 g of iron 9 nitrate 2 of 99% industrial strength, 308 g of zinc 6 nitrate of 95% industrial strength and 18.7 g of titanium tetrachloride of industrial strength are dissolved in 400 ml of propylene glycol. This is agitated for 30 minutes and mixed well.

Next, it is mixed with an aqueous solution containing 1600 ml of water and 320 g of industrial strength urea, these are mixed together and agitated.

This is heated under agitation, the heating is carried out for 3 hours at 100 deg C. A hydrolyzed product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 1.0 micrometer, 108 g of the fine powder of average particle size of the powder of 0.04 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Al/Fe/Ti of 100/40/0.5/10.

The atom bonding state of the powder that is obtained is measured, - Ti - O - Zn - O - Al< bond (a special absorption of 650 cm<sup>-1</sup>, 740 cm<sup>-1</sup>) is recognized.

The spectral transmissivity measurement result is shown in figure 4.

Implementation example 26

152 g of an aqueous solution of aluminum 9 nitrate of 97% industrial strength, 2.24 g of an aqueous solution

cerium 8 sulfate 1, 308 g of an aqueous solution of zinc 6 nitrate of 95% industrial strength and 18.7 g of titanium tetrachloride of industrial strength are dissolved in 1200 ml of isopropanol. This is agitated for 30 minutes and mixed well.

Next, it is mixed with an aqueous solution containing 1600 ml of water and 320 g of industrial strength urea, these are mixed together and agitated.

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This is heated under agitation, the heating is carried out for 4 hours at 80 deg C. Then, it is heated for 1 hour at 100 deg C. A hydrolyzed product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 0.5 micrometer, 109 g of the fine powder of average particle size of the powder of 0.05 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Al/Ce/Ti of 100/40/0.5/10.

The atom bonding state of the powder that is obtained is measured, - Ti - O - Zn - O - Al< bond (a special absorption of 650 cm-1, 740 cm-1) is recognized.

The spectral transmissivity measurement result is shown in figure 2.

#### Implementation example 27

4.72 g of iron 9 nitrate 2 of 99% industrial strength, 2.24 g of cerium 8 sulfate 1, 362 g of zinc 6 nitrate of 95% industrial strength and 32.9 g of titanium isopropoxide 32 made by Nihon Co. are dissolved in 1200 ml of isopropanol. This is agitated for 30 minutes and mixed well.

Next, it is mixed with an aqueous solution containing 1600 ml of water and 255 g of industrial strength urea, these are mixed together and agitated.

This is heated under agitation, the heating is carried out for 4 hours at 80 deg C. It is heated further for 1 hour at 100 deg C. A hydrolyzed product is obtained.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 0.5

micrometer, 105 g of the fine powder of average particle size of the powder of 0.03 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Fe/Ce/Ti of 100/1/0.5/10.

The atom bonding state of the powder that is obtained is measured, - Zn - O - Ti < bond (a special absorption of 600 cm<sup>-1</sup>, 740 cm<sup>-1</sup>) is recognized.

The spectral transmissivity measurement result is shown in figure 4.

#### Implementation example 28

152 g of an aqueous solution of aluminum 9 nitrate of 97% industrial strength, 2.14 g of an aqueous solution cerium 8 sulfate 1, 308 g of an aqueous solution of zinc 6 nitrate of 95% industrial strength and 2.01 g of an aqueous solution of iron 9 nitrate 2 of 99% industrial strength, 2.14 g of cerium 6 nitrate 1 aqueous solution and 18.7 g of titanium tetrachloride of industrial strength are dissolved in 400 ml glycerin. This is agitated for 30 minutes and mixed well.

Next, it is mixed with an aqueous solution containing 1600 ml of water and 322 g of industrial strength urea, these are mixed together and agitated.

This is heated under agitation, the heating is carried out for 3 hours at 100 deg C.

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 1 micrometer, 109 g of the fine powder of average particle size of the powder of 0.05 micrometers are obtained.

The composition of this powder product is analyzed and the result is an atom ratio of Zn/Al/Fe/Ce/Ti of 100/40/0.5/0.5/10.

The atom bonding state of the powder that is obtained is measured, - Ti - O - Zn - O - Al < bond (a special absorption of 650 cm-1, 740 cm-1) is recognized.

The spectral transmissivity measurement result is shown in figure 4.

Implementation example 29

34.4 g of an aqueous solution of aluminum nitrate of 97% industrial strength, 8.15 g of an aqueous solution of iron nitrate 2 of 99% industrial strength, 62.6 g of zinc nitrate aqueous solution of 95% industrial strength and 25.3 g of an aqueous solution of chromium nitrate 2, 6.4 of titanyl sulfate of industrial strength are dissolved in 500 ml water. This is agitated for 30 minutes and mixed well.



Next, it is mixed with an aqueous solution containing 1100 ml of water and 280 g of industrial strength urea and 1200 ml of ethanol, these are mixed together and agitated.

This is heated under agitation, the heating is carried out for 5 hours at 80 deg C. It is heated further for 1 hour at 100 deg C. A hydrolyzed product is obtained /12

This is filtered and washed with 1000 ml of water. It is dried at 150 deg C and pulverized with a hammer mill and a vibration mill.

Next, it is baked for 1 hour at a temperature of about 850 deg C, all the particle size is not larger than 1 micrometer, 101 g of the fine powder of average particle size of the powder of 0.08 micrometers are obtained.

The composition of this powder product is analyzed and these bondings are recognized - Zn - o - Al < bond, - Zn - o - Fe < bond, - Zn - O - Cr < bond, - Zn - O - Ce < bond and - Zn - O - Ti < bond.

The result from the analysis of the composition of this powder is the atomic ratio of Zn/Al/Fe/Cr/Ce/Ti is 100/50/10/30/200/20.

The spectral transmissivity measurement result is shown in figure 4.

### Implementation example 30

32.2 g of zirconium oxychloride and each of the raw material salts from Implementation example 29 are dissolved in 500 ml of water, mixed and agitated for 30 minutes.

Next, it is mixed with an aqueous solution containing 1100 ml of water and 310 g of industrial strength urea and 1200 ml of ethanol is added, these are mixed together and agitated.

The reaction, the filtering, the drying, the pulverizing and baking are performed similar to Implementation example 29, no particles are found to be larger than 0.05 micrometer, 111 g of fine powder of particle size of 0.01 micrometer is obtained.

The composition of this powder product is analyzed and these bonds are recognized - Zn - o - Al < bond, - Zn - o - Fe < bond, - Zn - O - Cr < bond, - Zn - O - Ce < bond, - Zn - O - Ti < bond and - Zn - O - Zr.

The result from the analysis of the composition of this powder is the atomic ratio of Zn/Al/Fe/Cr/Ce/Ti/Zr is 100/50/10/30/200/20/50.

The spectral transmissivity measurement result is shown in figure 4.

### Comparison example 1

23.9 g of aluminum oxide powder of industrial strength made by Wako Pure Chemicals and 76.1 g of Zinc oxide made by Yamada Pharmaceutical Industries are pulverized and mixed with a pore mill for 6 hours, it is baked for 1 hour at a temperature of about 700 deg C.

The atomic ratio Zn/Al of the powder that is obtained is 100/50 and the particle size is about 0.2 micrometer.

When analyzing the atomic bonding state of this powder, the bond - Zn - O - Al < was not recognized, the powder mixture consists simply of zinc oxide and aluminum oxide.

The spectral transmissivity is measured and the result is shown in figure 5.

#### Comparison example 2

9.24 g of titanium oxide powder of industrial strength made by Wako Pure Chemicals and 94.1 g of Zinc oxide made by Yamada Pharmaceutical Industries are pulverized and mixed with a pore mill for 6 hours, it is baked for 1 hour at a temperature of about 850 deg C.

The atomic ratio Zn/Ti of the powder that is obtained is 100/50 and the particle size is about 0.2 micrometer.

When analyzing the atomic bonding state of this powder, the bond - Zn - O - Ti < was not recognized, the

powder mixture consists simply of zinc oxide and aluminum oxide.

The spectral transmissivity is measured and the result is shown in figure 5.

#### Comparison example 3

9.24 g of titanium oxide powder of industrial strength made by Wako Pure Chemicals and 23.6 g of aluminum oxide made by the same Company and 94.1 g of zinc oxide of industrial strength made by Yamada Pharmaceutical Industries are pulverized and mixed with a pore mill for 6 hours, it is baked for 1 hour at a temperature of about 850 deg C.

The atomic ratio Zn/Al/Ti of the powder that is obtained is 100/40/10 and the particle size is about 0.2 micrometer.

When analyzing the atomic bonding state of this powder, the bond - Ti - O - Zn - O - Al < was not recognized, the powder mixture consists simply of zinc oxide and aluminum oxide.

The spectral transmissivity is measured and the result is shown in figure 5.

#### Comparison example 4

32.9 g of iron oxide powder of industrial strength made by Nihonben and 67.1 g of zinc oxide of industrial

strength made by Yamada Pharmaceutical Industries are pulverized and mixed with a pore mill for 6 hours, it is baked for 1 hour at a temperature of about 600 deg C.

The atomic ratio Zn/Fe of the powder that is obtained is 100/50 and the particle size is about 0.2 micrometer./13

When analyzing the atomic bonding state of this powder, the bond - Zn - O - Fe - < was not recognized, the powder mixture consists simply of zinc oxide and iron oxide.

The spectral transmissivity is measured and the result is shown in figure 5.

Comparison example 5

0.022 g of cerium oxide powder of industrial strength made by Wako Pure Chemicals and 102 g of zinc oxide of industrial strength made by Yamada Pharmaceutical Industries are pulverized and mixed with a pore mill for 6 hours, it is baked for 1 hour at a temperature of about 600 deg C.

The atomic ratio Zn/Ce of the powder that is obtained is 100/0.01 and the particle size is about 0.2 micrometer.

The spectral transmissivity is measured and the result is shown in figure 5.

#### Comparison example 6

1 g of iron oxide powder of industrial strength made by Nihonben, 6.37 g of aluminum made by Wako Pure Chemicals and 102 g of zinc oxide of industrial strength made by Yamada Pharmaceutical Industries are pulverized and mixed with a pore mill for 6 hours, it is baked for 1 hour at a temperature of about 600 deg C.

The atomic ratio Zn/Al/Fe of the powder that is obtained is 100/10/1 and the particle size is about 0.2 micrometer.

The spectral transmissivity is measured and the result is shown in figure 5.

#### Comparison examples 7, 8, 9, 10, 11, 12

The spectral transmissivity is measured for each of the powder, the aluminum oxide powder, the zinc oxide powder, the titanium oxide powder sold commercially, the iron oxide powder made by Nihonben, the titanium oxide fine particles made by Dexa that are used in Comparison examples 1 - 6, the results are shown in figure 5.

Each of the particle size of each oxide used in the comparison examples are shown in Table 1.

Table 1

| 比較例 | 酸化物                              | 粒 度                              |
|-----|----------------------------------|----------------------------------|
| 7   | 酸化チタン                            | 約 0.5 $\mu$ m ~ 約 5 $\mu$ m      |
| 8   | 酸化亜鉛                             | 約 0.5 $\mu$ m ~ 約 1 $\mu$ m      |
| 9   | 酸化鉄                              | 約 0.2 $\mu$ m ~ 約 1 $\mu$ m      |
| 10  | 酸化Fe <sub>2</sub> O <sub>3</sub> | 約 0.2 $\mu$ m ~ 約 1 $\mu$ m      |
| 11  | 酸化Fe <sub>2</sub> O <sub>3</sub> | 約 0.002 $\mu$ m ~ 約 0.05 $\mu$ m |
| 12  | 酸化チタン                            | 約 0.2 $\mu$ m ~ 約 2 $\mu$ m      |

Column 1: Comparison examples - 7 - 12

Column 2: Oxide - aluminum oxide; zinc oxide; iron oxide; titanium oxide; titanium oxide, titanium oxide

Column 3: Particle size - about 0.5 micrometer - about 5 micrometers; about 0.5 micrometer - about 1 micrometer; about 0.2 micrometer - about 1 micrometer; about 0.2 micrometer - about 1 micrometers; about 0.002 micrometer - about 0.05 micrometer; about 0.2 micrometer - about 2 micrometers

Comparison example 13

Instead of 1200 ml of ethanol used in Implementation example 1, 1200 ml of water is used, the hydrolysis is carried out, the drying and baking are performed.

The result is 98.2 g of powder of average particle size of 1 micrometer.

The composition of this powder has the atomic ratio of Zn/Al of 100/10.

When analyzing the atomic bonding state of this powder, the bond - Zn - O - Al < was recognized (the special absorption is  $650\text{cm}^{-1}$ ).

The spectral transmissivity is measured and the result is shown in figure 5. Furthermore, as a result of checking the catalytic activity of the raw material oxide powder used in the comparison examples and the fine complex oxides that are obtained, only the ultrafine titanium oxide particle made by Dexa displayed high level of catalyst activity. The film formation was difficult and in particular, the catalyst activity was not recognized in the others.

Brief explanation of the diagrams

Figures 1 - 4 show the spectral transmissivity line diagram in the ultraviolet - visible ray range of the metal oxide powder obtained from Implementation examples 1- 30.

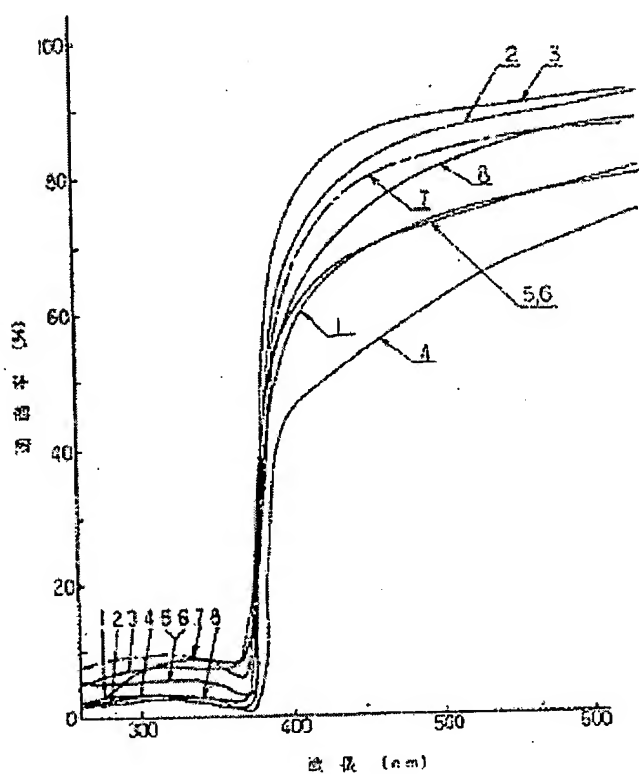
Figure 5 shows the spectral transmissivity curve diagram in the ultraviolet - visible ray region of the metal oxide powder treated in comparison examples 1 - 6, the powder of the complex metal oxide obtained in



comparison example 13 and the metal oxide powder obtained in comparison examples 1 - 6. The number in the diagram corresponds to the comparison number.

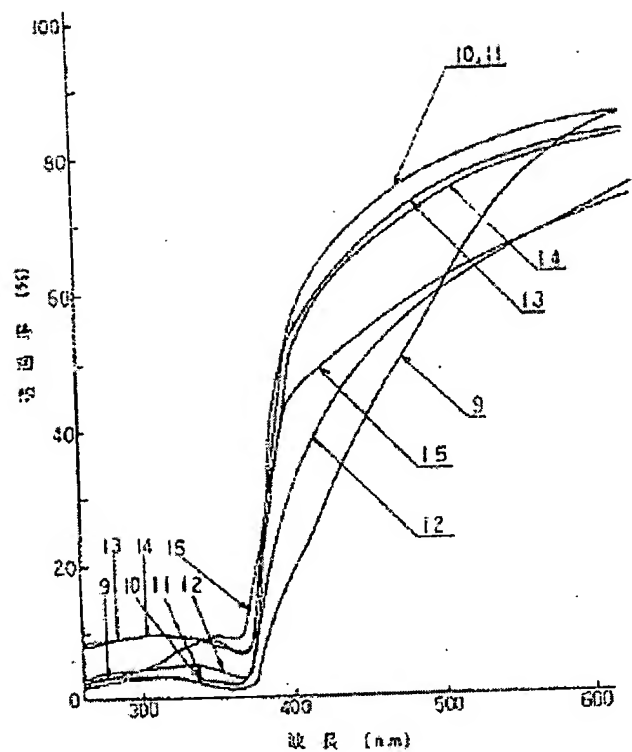
/14

Figure 1



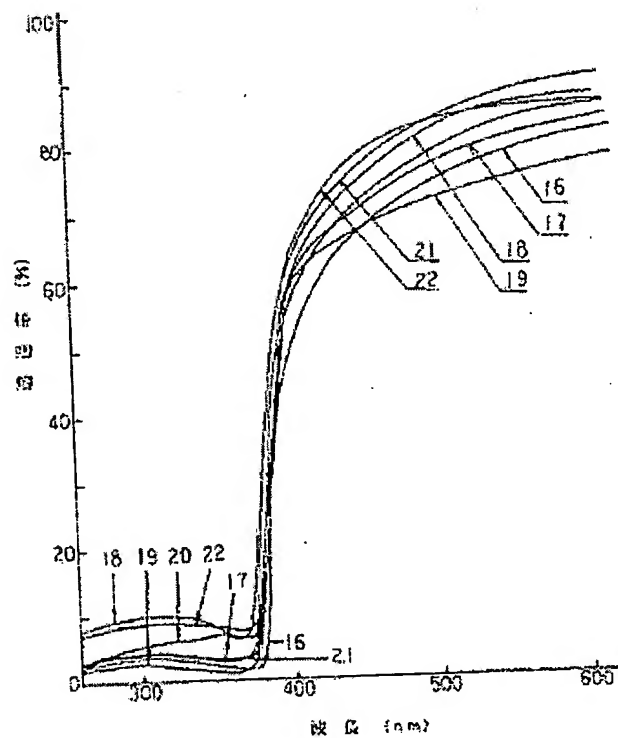
Spectral transmissivity (%) Vs wavelength (nm)

Figure 2



Spectral transmissivity (%) Vs wavelength (nm)

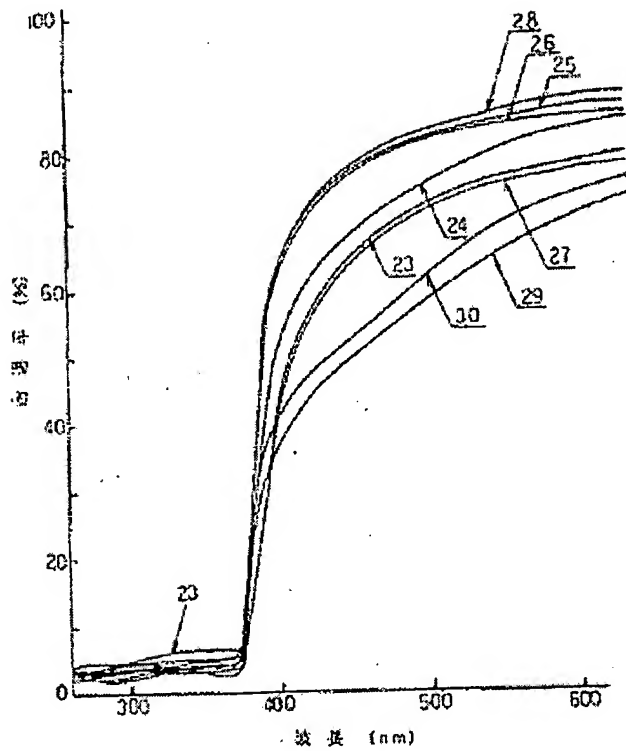
Figure 3



Spectral transmissivity (%) Vs wavelength (nm)

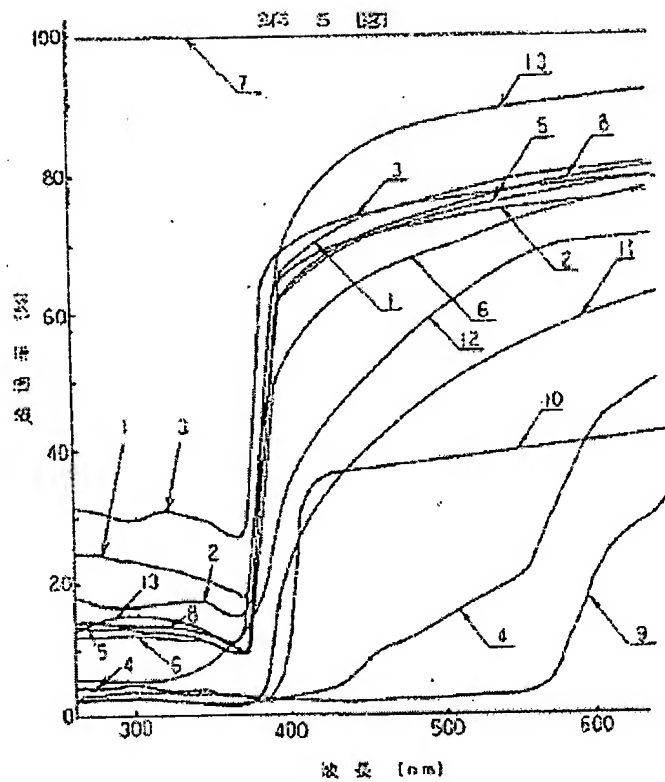
/15

Figure 4



Spectral transmissivity (%) Vs wavelength (nm)

Figure 5



Spectral transmissivity (%) Vs wavelength (nm)